Supplementary Information

Magnetic relaxations in four-coordinate Dy(III) complexes: effects of anionic surroundings and short Dy–O bond

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Table S1: Selected bond lengths (Å) and angles (deg) in complex **1**[a]

Dy(1)-N(1)	2.2948(15)	N(2)-Dy(1)-N(1)	93.84(6)
Dy(1)-N(2)	2.2824(16)	N(1)#1-Dy(1)-N(1)	103.84(7)
Li(1)-O(1)	1.941(3)	N(2)#1-Dy(1)-N(1)	134.09(6)
Li(1)-O(2)	1.930(4)		

[a] Symmetry codes: #1 -x,y,-z+1/2

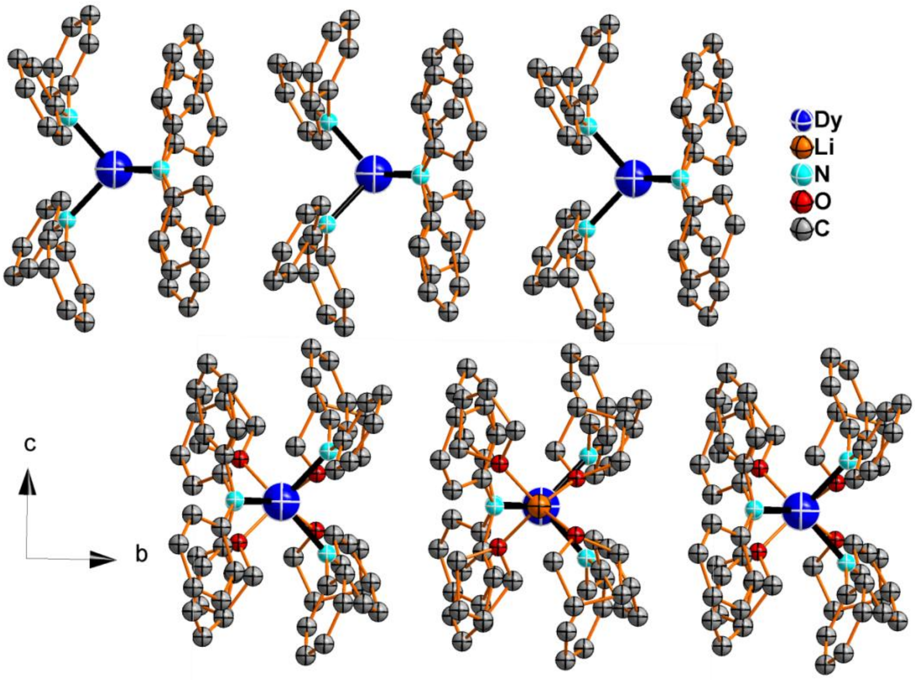

Fig. S1 Packing diagram for complex **1** shown along the crystallographic a axis

Table S2: Selected bond lengths (Å) and angles (deg)in complex **2**

Dy(1)-O(2)	2.068(3)	O(2)-Dy(1)-O(3)	107.94(12)
Dy(1)-O(3)	2.152(3)	O(2)-Dy(1)-O(1)	109.78(13)
Dy(1)-O(1)	2.195(3)	O(3)-Dy(1)-O(1)	78.04(11)
Dy(1)-N(1)	2.273(4)	O(2)-Dy(1)-N(1)	112.76(15)
Li(1)-O(4)	1.893(10)	O(3)-Dy(1)-N(1)	115.25(14)
Li(1)-O(1)	1.898(9)	O(1)-Dy(1)-N(1)	127.30(14)
Li(1)-O(3)	1.908(10)		

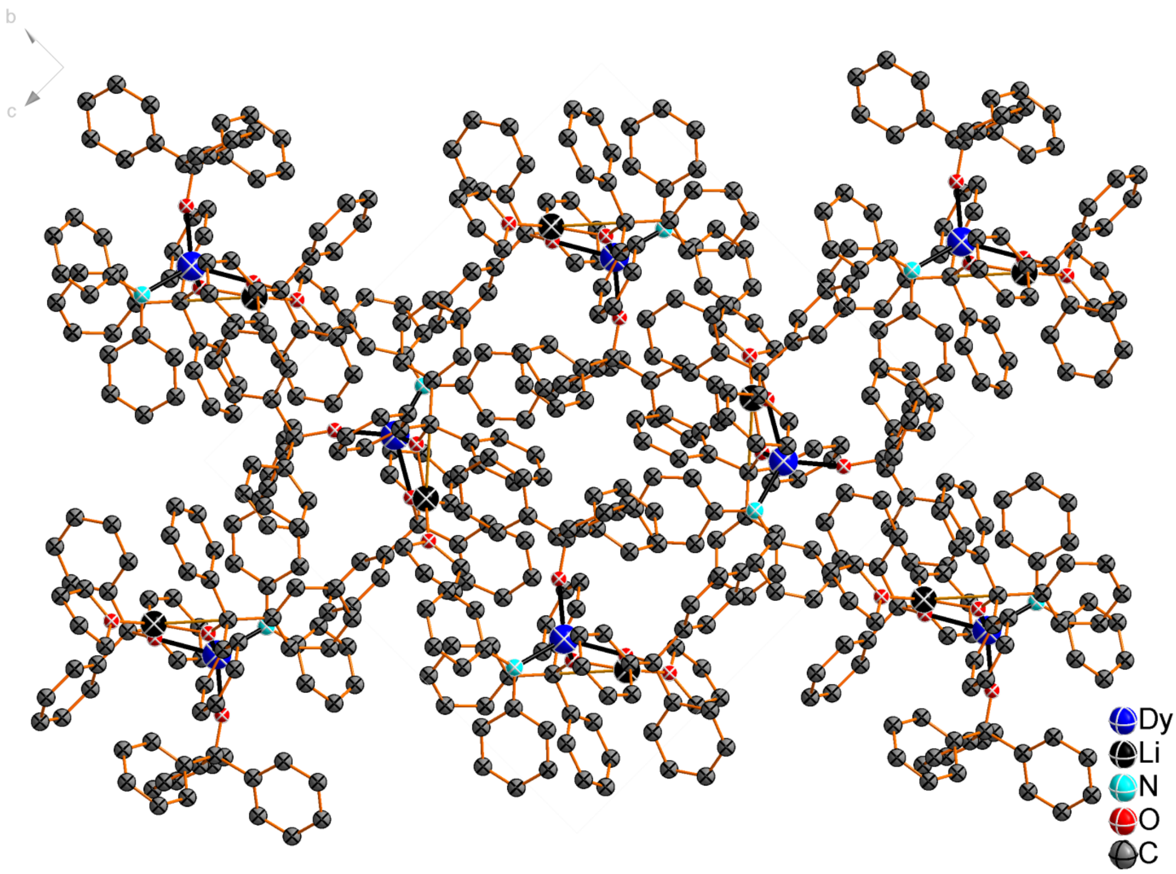

Fig. S2 Packing diagram for complex **2** shown along the crystallographic *a* axis

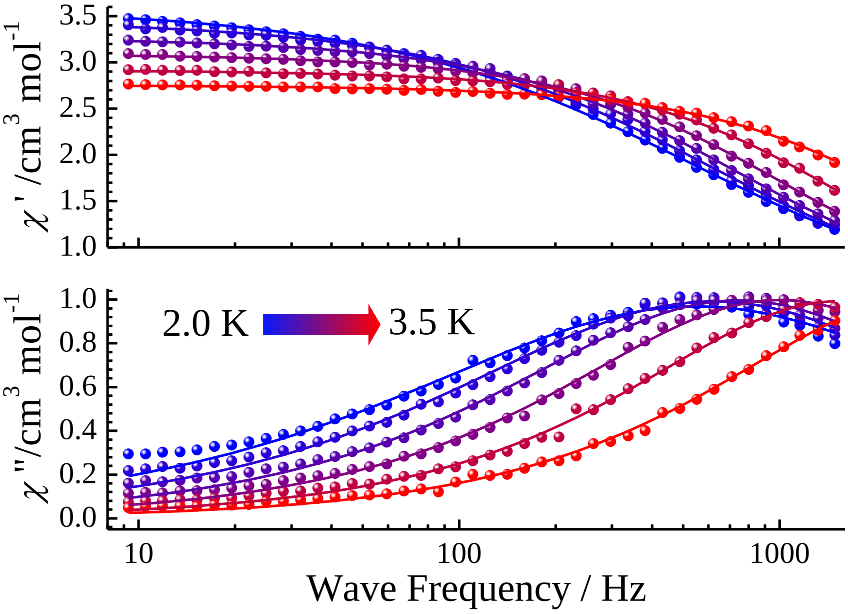

Fig S3, Frequency-dependence of the χ' and χ'' ac susceptibility signals under 1600 Oe dc field at the temperature from 2.0 K (blue) to 3.5 K (red) for **1**. The solid lines are best fits.

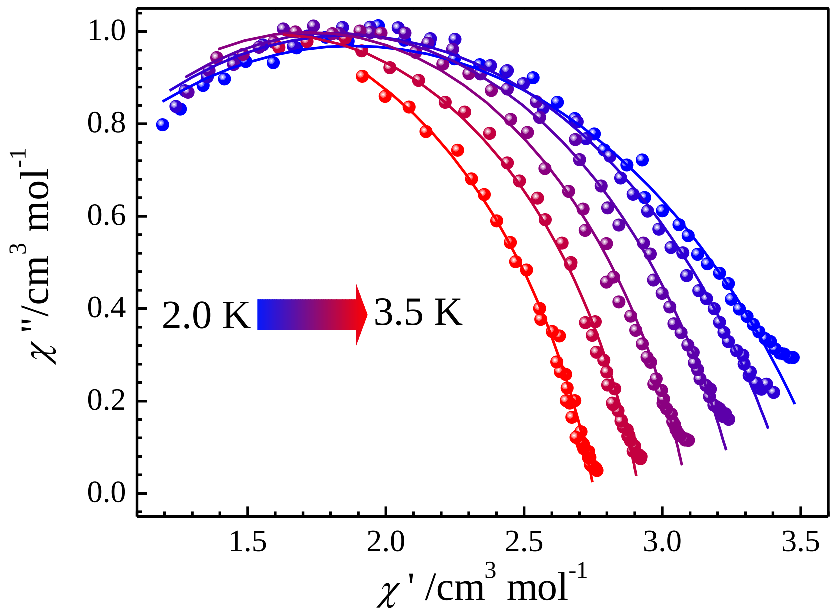

Fig. S4 Cole-Cole plots using the frequency-dependence ac susceptibility data under the dc field of 1600 Oe for **1** at the indicated temperature. The solid lines are the best fits obtained with a generalized Debye model (with α values ranging from 0.17 to 0.35).

Table S3: Relaxation Fitting Parameters of a generalized Debye model for **1**.

T	χ_S	χ_T	τ	α
2.0	0.17014	3.60646	2.92295E-4	0.34573
2.3	0.27802	3.4538	2.58967E-4	0.28871
2.6	0.36647	3.26714	2.15726E-4	0.2333
2.9	0.35227	3.08952	1.59495E-4	0.19748
3.2	0.28526	2.91563	1.03616E-4	0.17671
3.5	0.02544	2.75317	5.4296E-5	0.18724

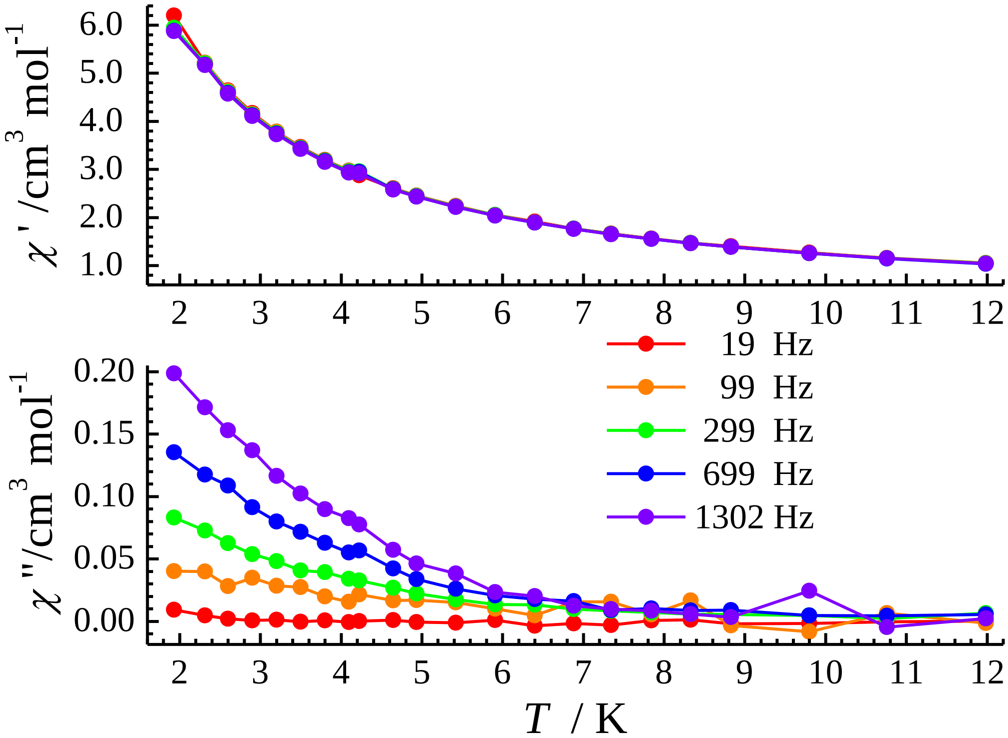

Fig S5, Temperature-dependence of the χ' and χ'' susceptibility signals under zero dc field at the indicated frequencies for complexes **2**. The solid lines are guides for vision.

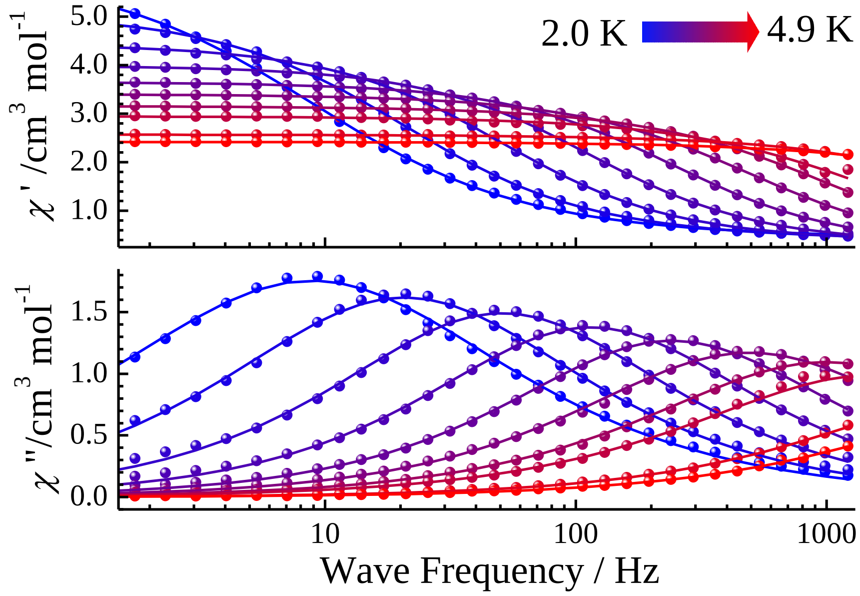

Fig S6, Frequency-dependence of the χ' and χ'' ac susceptibility signals under 700 Oe dc field at the temperature from 2.0 K (blue) to 4.9 K (red) for **2**. The solid lines are best fits.

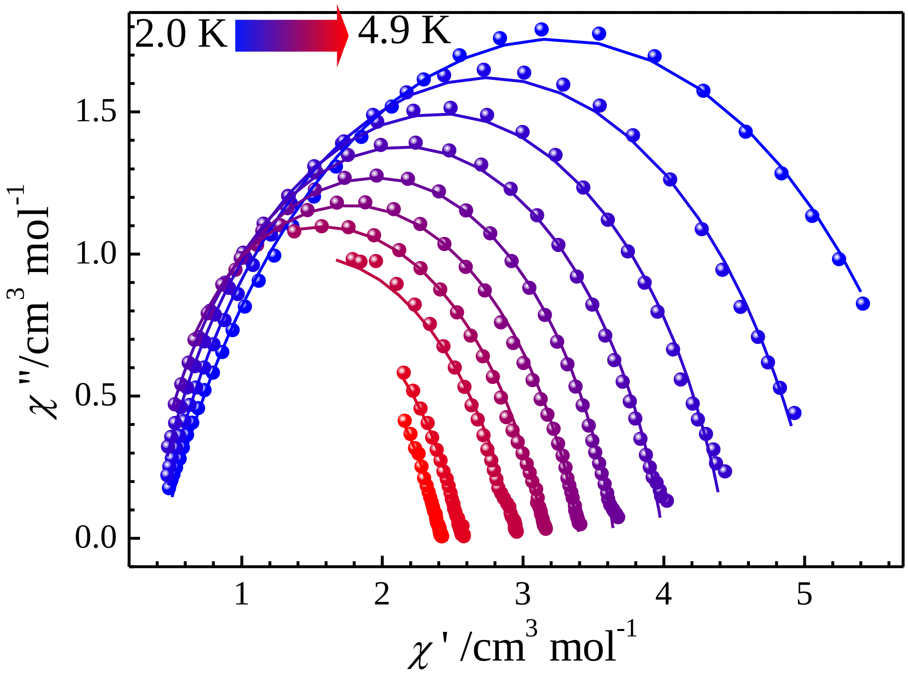

Fig. S7 Cole-Cole plots using the frequency-dependence ac susceptibility data under a dc field of 700 Oe for **2** from 2.0 K (red) to 4.9 K (blue). The solid lines are the best fit obtained with a generalized Debye model (with α ranging from 0.18 to 0.26).

Table S4: Relaxation Fitting Parameters of a generalized Debye model for **2**.

T	χ_S	χ_T	τ	α
2	0.43033	6.02086	0.01807	0.28576
2.3	0.40133	5.09638	0.00767	0.2308
2.6	0.35531	4.4449	0.00314	0.19668
2.9	0.30382	3.9963	0.00139	0.18338
3.2	0.25141	3.65019	6.59464E-4	0.18312
3.5	0.13421	3.40108	3.23366E-4	0.20748
3.8	4.57536E-8	3.15486	1.59538E-4	0.22593
4.1	8.29091E-8	2.94301	8.74393E-5	0.23581
4.6	1.06789E-7	2.56831	2.91366E-5	0.24614
4.9	1.68694E-7	2.41899	1.77789E-5	0.25584

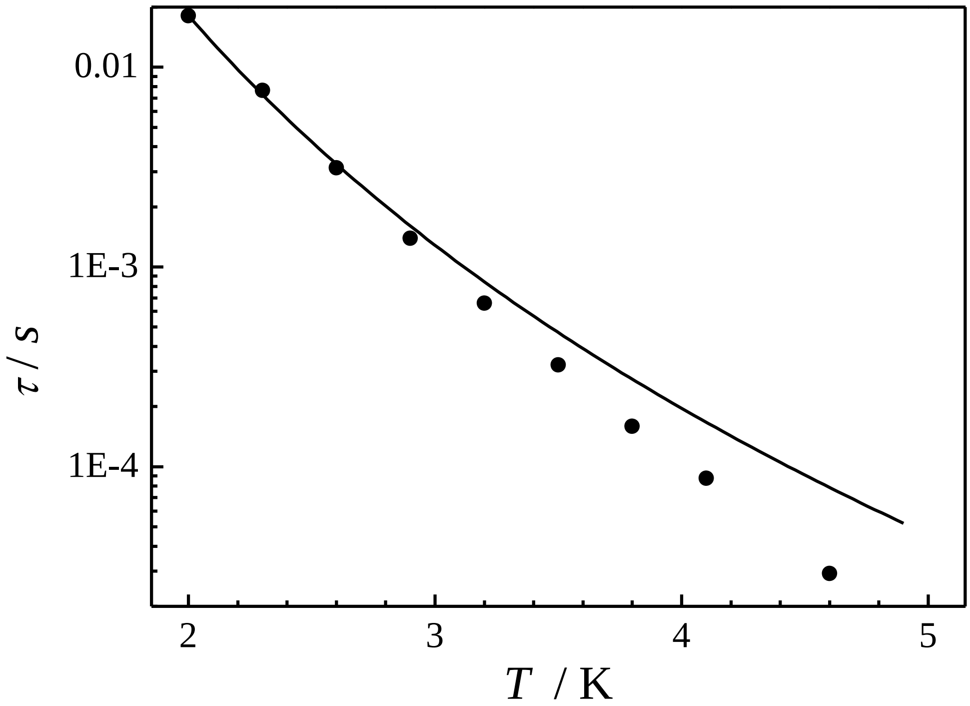

Fig.S8 Plots of τ versus T for **2**.The solid lines are best Raman fit $\tau^{-1} = cT^{n}$ with c = 0.59 K^{-n} s^{-1} and n = 6.53.

Table S5 Energy levels and eigenstates for **1** obtained from fitting.

Energy / cm^{-1}	Eigenstate
0	69.8%$\vert$ ± 15/2> + 30.1%$\vert$ ± 7/2> + 0.1%$\vert$ ± 1/2>
24.46	69.6%$\vert$ ± 7/2> + 30.1%$\vert$ ± 15/2> + 0.3%$\vert$ ± 1/2>
35.02	95.7%$\vert$ ± 5/2> + 4.0%$\vert$ ± 3/2>+ 0.3%$\vert$ ± 13/2>
73.17	100%$\vert$ ± 9/2>
97.07	95.4%$\vert$ ± 3/2> + 4.0%$\vert$ ± 5/2> + 0.6%$\vert$ ± 11/2>
139.9	99.6%$\vert$ ± 1/2> + 0.4%$\vert$ ± 7/2>
195.6	99.4%$\vert$ ± 11/2> + 0.6%$\vert$ ± 3/2>
269.1	99.7%$\vert$ ± 13/2> + 0.3%$\vert$ ± 5/2>

Table S6 Energy levels and eigenstates for **2** obtained from fitting.

Energy / cm^{-1}	Eigenstate
0	92.0%$\vert$ ± 15/2> + 7.8%$\vert$ ± 9/2> + 0.2%$\vert$ ± 3/2>
59.9	97.1%$\vert$ ± 13/2>+ 2.5%$\vert$ ± 7/2> + 0.3%$\vert$ ± 1/2> + 0.1%$\vert$ ± 5/2>
160.0	80.3%$\vert$ ± 11/2> + 14.6%$\vert$ ± 1/2> + 3.1%$\vert$ ± 5/2>+ 2.0%$\vert$ ± 7/2>
217.9	47.4%$\vert$ ± 9/2> + 47.3%$\vert$ ± 3/2> + 5.3%$\vert$ ± 15/2>
230.5	47.9%$\vert$ ± 1/2> + 36.7%$\vert$ ± 7/2> + 15.1%$\vert$ ± 11/2>…
277.5	69.5%$\vert$ ± 5/2> +17.7%$\vert$ ± 7/2>…
361.4	52.5%$\vert$ ± 3/2> + 44.8%$\vert$ ± 9/2>
411.1	41.2%$\vert$ ± 7/2> + 30.3%$\vert$ ± 1/2> + 27.2%$\vert$ ± 5/2>